

Thermoelectric response near a quantum critical point: the case of CeCoIn₅

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We present a study of thermoelectric coefficients in CeCoIn₅ down to 0.1 K and up to 16 T in order to probe the thermoelectric signatures of quantum criticality. In the vicinity of the field-induced quantum critical point, the Nernst coefficient ν exhibits a dramatic enhancement without saturation down to lowest measured temperature. The dimensionless ratio of Seebeck coefficient to electronic specific heat shows a minimum at a temperature close to threshold of the quasiparticle formation. Close to $T_c(H)$, in the vortex-liquid state, the Nernst coefficient behaves anomalously in puzzling contrast with other superconductors and standard vortex dynamics.

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CeCoIn₅ is an unconventional superconductor with an intriguing normal state[1]. Its behavior is peculiar near the upper critical field, where the energy scale governing various electronic properties is vanishingly small and increases with increasing magnetic field[2, 3], a behavior expected in presence of a Quantum Critical Point(QCP)[4]. The proximity of this QCP to the upper critical field in CeCoIn₅ is puzzling[5, 6, 7]. The possible existence of a FFLO state[8] and/or an elusive magnetic order are a subject of recent intense research. On the other hand, even in the absence of magnetic field, the normal state presents strong deviation from the standard Fermi-liquid behavior[1, 9]. The application of pressure leads to the destruction of superconductivity and the restoration of the Fermi liquid[6, 10, 11]. The link between the field-induced and the pressure-induced routes to the Fermi liquid is yet to be clarified.

During the last three years, the anomalous properties of CeCoIn₅ near the field-induced QCP have been reported thanks to measurements of specific heat[3], electric resistivity[2], thermal transport[7] and Hall effect[13]. In this paper, new insight on the quantum criticality is given via thermoelectric response down to 0.1K. As far as we know, this is the first experimental investigation of the thermoelectric tensor in the vicinity of a QCP, a subject of several theoretical studies[16, 17, 18]. Single crystals were grown by self-flux method. Thermoelectric coefficients were measured with one heater and two RuO₂ thermometers in magnetic field along c -axis. The heat current was applied along the basal plane. Previous studies of thermoelectricity in CeCoIn₅ detected a large Nernst coefficient and a field-dependent Seebeck coefficient in the non-Fermi liquid regime above T_c [14] and an additional field scale at 23 T[15]. Here, we find that the most spectacular thermoelectric signature of quantum criticality is a drastic enhancement of the Nernst coefficient, ν . The vanishingly small Fermi energy, which was previously detected by a nearly diverging enhancement of the A coefficient of resistivity ($\rho = \rho_0 + AT^2$)[2] and the

Sommerfeld coefficient of specific heat ($\gamma = C_{el}/T$)[3], leads also to an apparently diverging ν/T . These results show two distinct anomalies close to $H_{c2}(0)$ and $T_c(0)$ which are different in the origin. This conclusion cannot be derived from other probes mentioned above. We also find a milder enhancement of the Seebeck coefficient near the QCP. Moreover, the ratio of thermopower to electronic specific heat, expressed in appropriate units[19], remains close to unity even in the vicinity of the QCP. The temperature dependence of this ratio presents a minimum at a temperature roughly marking the formation of well-defined quasi-particles[7].

Figure 1 presents the data obtained by measuring the Nernst and the Seebeck coefficients at various magnetic fields. Since the thermoelectric response of Fermions is expected to be T -linear well below their Fermi temperature, what is plotted in the figure is the temperature dependence of the two coefficients divided by temperature. As seen in fig. 1(a), the Seebeck coefficient, S vanishes in the superconducting state. In the normal state, S/T increases with decreasing temperature for all fields. For fields exceeding 5.4 T, the normal state extends down to zero temperature and a finite S/T in the zero-temperature limit can be extracted. For a field of 16 T (which is well above the quantum critical region) S/T saturates to a value of about $13 \mu\text{VK}^{-2}$. For fields between 5.4 T and 16 T, S/T presents a non-monotonous temperature dependence. An upturn below 0.15 K is visible for $\mu_0 H \simeq 5.5$ T (i.e. in the vicinity of the QCP) curves. Note that this upturn leads to a moderate enhancement of S/T . The overall change in the magnitude of S/T is about 70 %. On the other hand, the temperature dependence of the Nernst coefficient divided by temperature ν/T reveals a more dramatic signature of Quantum criticality. As seen in figure 1(b), for $\mu_0 H = 5.5$ T and $\mu_0 H = 6$ T, below 1 K, ν/T is steadily increasing with decreasing temperature. No such enhancement occurs for $\mu_0 H = 16$ T, far above QCP. At the lowest measured temperature (~ 0.1 K), ν/T is five-fold en-

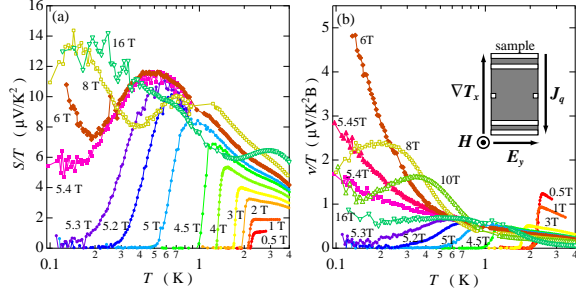


FIG. 1: (a) The Seebeck coefficient divided by temperature as a function of temperature for different magnetic fields in a semi-log plot. Note the upturn near the QCP. (b) Temperature dependence of the Nernst coefficient divided by temperature ν/T . Close to the QCP, this quantity never saturates. The inset defines the convention used for the sign of the Nernst coefficient (see text).

hanced near the QCP (~ 6 T) compared to its 16 T value. Since the thermal Hall conductivity κ_{xy} in CeCoIn₅ becomes large at low temperatures due to enhancement of the mean-free-path of the electrons [12], the transverse thermal gradient $\nabla_y T$ could generate a finite transverse electric field E_y . Therefore, the (measured) adiabatic and the (theoretical) isothermal Nernst coefficients are not identical in CeCoIn₅. However, using the value of $|\nabla_y T|/|\nabla_x T| \sim 0.1$ at 5.2 T reported in Ref. [12], the difference between these two is estimated to be about 10 %, indicating that the observed enhancement is not due to a finite $\nabla_y T$. We will argue below that this enhancement reflects a concomitant decrease in the magnitude of the normalized Fermi energy as previously documented by specific heat and resistivity measurements.

The thermoelectric response of CeCoIn₅ in the vicinity of QCP can be better understood by complementing our data with the information extracted by other experimental probes[2, 3], which originally detected a quantum critical behavior near H_{c2} . In particular, an interesting issue to address is the fate of the correlation observed between thermopower and specific heat of many Fermi liquids in the zero-temperature limit. In a wide range of systems, the dimensionless ratio linking these two is of the order of unity ($q = \frac{SN_A e}{T\gamma} \simeq \pm 1$, with $\gamma = C_{el}/T$, N_A the Avogadro number and e the charge of electron)[19]. What happens to such a correlation at a quantum critical point? Combining the specific heat data reported by Bianchi *et al.*[3] with our thermopower results allows us to address these questions. Fig. 2(a) presents q computed in this way as a function of temperature. The first feature to remark is that q remains of the order of unity even in the quantum critical region. Note that, theoretically, this correlation arises because S/T and γ are both inversely proportional to the normalized Fermi energy and thus q is expected to be of the order of (and *not* rigorously equal to) unity[17]. According to our result ($q \simeq 0.9$ at 6 T and 0.1 K), this correlation holds even when the normal-

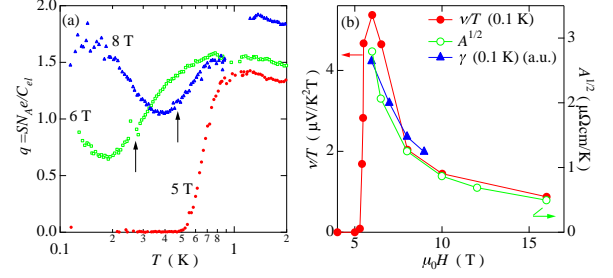


FIG. 2: (a) The temperature dependence of q , the dimensionless ratio of thermopower to electronic specific heat at three magnetic fields. The temperature marked by the arrow designs the threshold of the quasi-particle formation according to the temperature dependence of the Lorenz number as reported by Paglione *et al.*[7] (b) A comparison of the field dependence of ν/T , γ (as reported in ref. [3]) and $A^{1/2}$ (taken from ref. [2]).

ized Fermi Energy becomes vanishingly small. The second feature of interest in figure 2(a) is the temperature dependence of q , which presents a minimum. For both fields, the temperature at which this minimum occurs is close to the one where the Lorenz number ($L = \frac{\kappa}{\sigma T}$) linking thermal, κ , and electric, σ , conductivities present also a minimum. Paglione and co-workers, who report this latter feature, argue that this temperature marks the formation of well-defined quasi-particles[7]. This is a temperature below which both thermal and electric resistivities display a $T^{3/2}$ temperature dependence. Remarkably, Miyake and Kohno, who provided a theoretical framework in a periodic Anderson model for the correlation between thermopower and specific heat, predicted that q should deviate downward from unity in presence of an antiferromagnetic (AF) QCP leading to hot lines on the Fermi surface[17].

We now turn to the Nernst coefficient. In a simple picture, it is proportional to the energy derivative of the relaxation time at the Fermi energy[20]. In a first approximation, it tracks a magnitude set by the cyclotron frequency, the scattering time and the Fermi energy[21]. Since it scales inversely with the Fermi Energy, there is no surprise that it becomes large in heavy-fermion metals[14, 15] and in particular in heavy-Fermion semi-metals[22, 23], where both the heavy mass of electrons and the smallness of the Fermi wave-vector contribute to its enhancement ($\nu/T \propto 1/(k_F \epsilon_F)$). Now, since the Fermi energy (broadly defined as the characteristic energy scale of the system) becomes very small near a QCP, one would expect a large Nernst coefficient in agreement with the experimental observation reported here.

With these phenomenological considerations in mind let us compare the behavior of the Nernst coefficient with specific heat and resistivity. Both γ and A , the T^2 term of the resistivity ($\rho = \rho_0 + AT^2$) inversely scale with the Fermi Energy, ϵ_F . Therefore, both are enhanced when the Fermi energy is small. Since these two quantities are

linked by the Kadowaki-Woods relation ($\gamma^2 \propto A$), the enhancement is more pronounced in A than in γ . Figure 2(b) compares the field-dependence of $A^{1/2}$, γ and ν/T . In a naive picture, the enhancement of the three quantities are comparable in magnitude. This quantitative correlation suggests that the main reason for the enhancement of ν/T near QCP is due to a small ϵ_F . It is instructive to trace a contour plot of this quantity in the temperature-field plane. This is done in Fig. 3 with a logarithmic color scale in order to enhance the contrast. Note that contrary to the other probes, there is no need to subtract an offset from the Nernst data. In the case of specific heat, one should subtract the Schottky contribution at low temperature[3] and high-field, and the phonon contribution at high temperature. In the case of resistivity the T^2 behavior is interrupted at low temperature and high-fields by an upturn due to the temperature-dependent magnitude of $\omega_c\tau$ [2]. As seen in Fig. 3, ν/T becomes very large near the QCP, which constitutes the main hearth of the figure. However, there is a second one at zero field just above T_c , which was identified by previous measurements[14]. This zero-field hot region corresponds to a purely linear resistivity and anomalously enhanced Hall coefficient[9] due to strong anisotropic scattering by AF fluctuations[11], which can also enhance the Nernst coefficient[24]. On the other hand, close to the QCP, the magnitude of Hall coefficient[13] is comparable to its value at room-temperature or in LaCoIn_5 [11]. Therefore, there appears to be two distinct sources for the enhancement of the Nernst coefficient. In the zero-field regime just above T_c , it is enhanced mostly because of strong inelastic scattering associated with AF fluctuations, but in the zero-temperature regime just above H_{c2} , it becomes large because of the smallness of the Fermi energy. The occurrence of superconductivity impedes to explore the route linking together these two hot regions of the (B,T) plane. The inset of the figure compares the evolution of energy scales detected by different experimental probes near the QCP.

We now turn to the puzzling behavior of the Nernst coefficient in the vicinity of the superconducting transition. Deep into the superconducting state, there is no measurable Nernst signal, as illustrated by the existence of the black area in Fig. 3. On the other hand, close to $H_{c2}(T)$ (or alternatively, near $T_c(H)$), vortices can move and an additional contribution to the Nernst signal is expected. In the entire range of our study, the Nernst coefficient keeps the same sign which is presented in the inset of Fig. 1. Such a Nernst coefficient is negative according to a textbook convention on the sign of the thermoelectric coefficients[25]. However, the literature on the vortex Nernst effect[26] usually takes for positive the Nernst signal generated by vortices moving from hot to cold, which leads to an opposite convention. The sign of the Nernst effect in CeCoIn_5 is negative according to the textbook convention[25], but positive according to the vortex one[26, 27]. Indeed, contrary to quasi-particles, the Nernst signal produced by vortices

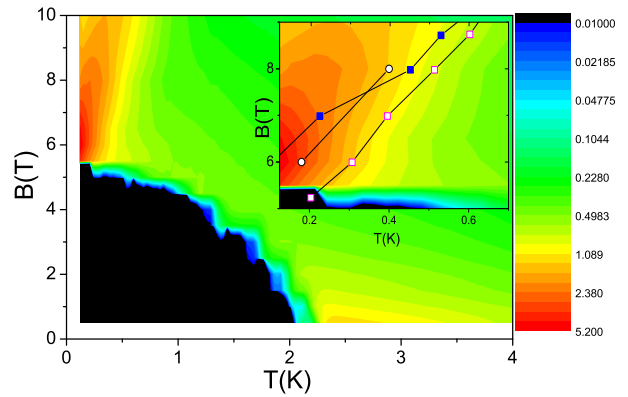


FIG. 3: Contour plot of ν/T in the (B,T) plane. The color scale is logarithmic. Note the presence of two hot regions close to H_{c2} and T_c . The inset is a zoom on the region near H_{c2} . The variation of three temperature scales, the onset of T^2 resistivity (solid squares), the minimum in L/L_0 (open squares) the and minimum in q (open circles) with magnetic field is also shown.

should have a fixed sign. A thermal gradient $\nabla_x T$ generates a force on a vortex because its core has an excess of entropy. The direction of this force is thus thermodynamically determined; vortices move along the thermal gradient from hot to cold region. The orientation of electric field is also unambiguously set by the direction of the vortex movement and the vortex Nernst signal is not expected to have an arbitrary sign. In order to separate the vortex and the quasi-particle contributions to the Nernst signal, we put under careful scrutiny the effect of superconducting transition on three coefficients : $\rho(T)$, $S(T)$ and $N(T)$. As illustrated in fig. 4(a) and 4(b), with the onset of superconductivity, the Nernst signal, N , collapses faster than both resistivity and the Seebeck coefficient. This robust feature was observed for *all* magnetic fields. On the other hand, the collapse in $\rho(T)$ and $S(T)$ closely track each other. This latter feature, which was also observed in cuprates[27], suggests that the Seebeck response is essentially generated by quasi-particles. Therefore, the most natural assumption regarding their contribution to the Nernst signal in the vortex liquid regime is that $N_{qp}(T)$ also follows $\rho(T)$ and $S(T)$ and the vortex contribution to the Nernst signal can be obtained by subtracting the normalized Seebeck coefficient off the normalized Nernst one. Fig. 4(c) and 4(d) show that this procedure clearly resolves a signal of opposite sign. Thus, the most straightforward interpretation of the faster collapse of $N(T)$ implies an additional source of Nernst signal in the vortex liquid regime with a sign *opposite* to the predominant one and also to the one expected for vortices moving along the heat flow.

This result appears incompatible with the standard picture of vortex dynamics driven by a thermal gradient. However, one shall not forget that additional forces on vortices besides thermal force may be present. CeCoIn_5

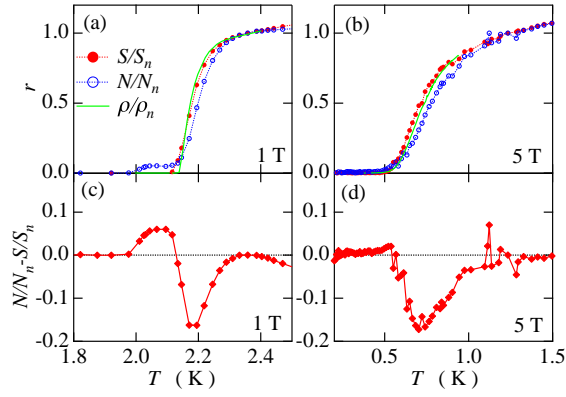


FIG. 4: (a)(b) Normalized magnitudes r of $r = N/N_n$ (open circles), S/S_n (solid circles) and ρ/ρ_n (solid line) in the vicinity of superconducting transition for 1 T and 5 T. For all fields, the onset of superconductivity leads to a faster collapse of the Nernst signal. Note also the small shoulder at 1 T. (c)(d) The additional contribution to transverse thermoelectricity in the vortex liquid regime obtained by subtracting the normalized Seebeck coefficient off the normalized Nernst signal.

is distinguished from other superconductors by the possible occurrence of an anti-ferromagnetic state in the normal core of its vortices. This feature could decrease the entropy excess of the vortices and reduce the intensity of the thermal force, which can therefore be vanquished by another source of vortex movement. As first noted

by Ginzburg[28], in a superconductor subject to a thermal gradient, a quasi-particle current (which carry heat) and a supercurrent (which does not) counterflow in order to keep the charge current zero[27]. In ordinary conditions, this counterflow generates a transverse Magnus force on vortices[29]. Its role in the context of superclean CeCoIn₅[12] needs an adequate theoretical treatment.

Another remarkable feature of Fig. 4 is the presence of a small shoulder in the temperature dependence of the Nernst effect at the end of the transition. The shoulder is present in an extended range of magnetic fields and only disappears in the proximity of H_{c2} . There seems to be a narrow temperature window, where a thermal gradient can create a transverse electric field, but a current does not produce any electric field. The simplest explanation for such a discrepancy would imply a threshold force to depin vortices, f_{dp} attained by the applied temperature gradient, but not by the applied current. However, this feature was found to be robust and no change was detected by modifying the magnitude of the applied thermal gradient. Clearly, the sign and the fine structure of the Nernst effect in the vortex liquid regime of CeCoIn₅ need further investigation.

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